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PATENT SPECIFICATION

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11

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(54) PROCESSES FOR PRODUCING VITREOUS FIBRES

We. JAPAN INORGANIC MÀTÉRIAL CO., LTD., a Japanese Body Corporate, of No. 1044-11, Fugisawa, Fugisawa-shi, Kanagawa-ken, Japan, do 5 nereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:-

This invention relates to processes for pro-

ducing vitreous fibres.

This invention provides a process for producing vitreous fibres, in which process a powder, comprising from 35 to 75% (cal-15 culated as silica) of a silicaceous material, from 10 to 55% (calculated as calcium oxide) of a calciferous material, minor amounts of metal oxides, and unavoidable impurities, is fused to form a melt which is passed through 20 orifices or spinnerers to form the desired vitreous fibres.

Thus, silica (SiO₂) and calcium oxide (CaO) are effectively the two principal ingredients of the powder, but it also contains smaller amounts of oxides, and by the term "minor amounts of metal oxides" is meant amounts of from 2 to 13% of alumina (Al₂O₂), from 0 to 10% of zirconium oxide ZrO,), from 0 to 4% of ferric oxide (Fe₂O₃), from 0 to 3% of sodium oxide (Na₂O), from 0 to 3% of potassium oxide (K₂O), from 0 to 2% of magnesium oxide MgO) and from 0 to 1% of titanium oxide (TiO₁).

All percentages quoted in the Specification

refer to percentages by weight.

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The powder could be made up by mixing together the required amounts of the various oxides — thus, by mixing suitable amounts of silica calcium oxide, alumina, ferric oxide and so on. However, many of the oxides themselves are very reactive, and as it is often most convenient to add the silicaceous material in the form of silicates and/or aluminosilicates. Such compounds are particularly convenient since they are widely available; a most suitable source of material for the powder used in the process of the invention is portland cement (known also as white cement),

which contains silicon, calcium, aluminium and iron in the following amounts (calculated, as are all percentage compositions quoted hereinafter, in terms of their oxide):—

SiO, 20 to 32%; 60 to 72%; Ca01 to 5%; and Al₂O₃ Fe₂O₃ 1 to 6%.

It is, therefore, preferable to make up the powder to be used in the process of the invention by mixing portland cement with a silicarich material to give the desired composition.

Portland cement (which is commonly termed simply "cement", and will be referred to as such hereinafter) is produced by grinding limestone and clay to a fine powder, forming this powder into a thick slurry with water, and firing it in a kiln. At a temperature of about 1500° C, the aluminosilicates from the clay are sintered to form small balls called clinkers. The clinkers are then crushed, for example in a ball mill, to give the fine powder known as cement. It is normal practice then add small amounts of gypsum (CaSO, 2H2O) to the cement - typically up to 2% of gypsum is added.

However, gypsum decomposes to give sulphur trioxide (SO₃) gas at the temperature used in vitreous fibre manufacture, and the production of any gas is undesirable since it may lead to the production of bubbles in the fibres. Consequently it is advantageous, though not essential, to use in the process of the invention cement containing substantially no gypsum or other material capable of releasing SO₃.

The advantages of using cement as a starting material to make vitreous fibres are:

a) it is readily available in large quantities all over the world;

b) it is stable if kept dry; c) it is available in the right form — a fine powder;

d) it is cheap. However, until now, it has proved virtually impossible to prepare vitreous fibres simply by heating cement in a manner analagous to

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the conventional preparations of vitreous fibres (in which a suitable glass is heated to from 1100 to 1500° C, and formed into the desired fibres). However, by the addition of silicarich material to the cement a powder may be formed from which vitreous fibres may be prepared. By the term "silica-rich material" as used herein is meant a material which contains at least 32.6% (calculated as silica) of silicon. Naturally, the higher the silica content of this silica-rich material, the smaller the amount that has to be added to the cement to give a powder having a silica content in the required range; therefore preferably the silica-rich material contains greater than 60% 15 (calculated as silica) of silicon. Silica itself can be used; commercial silica powder is readily available, and contains from 98 to 99% silica. However, there are many naturally-occurring silica-rich materials which may be used, and these are preferred because of their lower cost and ready availability. Examples of such materials are silica sand (97 to 98% silica), certain white clays (approximately 95% silica), shirasu (SiO2 65 to 73%, Al2O3 12 to 16%, Fe₂O₃ 1 to 3%, CaO 2 to 4%, K₂O 2 to 4%, Na₂O 3 to 4%), which is available in large quantities in Kagoshima Prefecture, Japan, and zircon flour (SiO₂ 30 32.60%, ZrO₂ 66.90%, TiO₂ 0.12%, Al₂O₃ 0.43%, Fe₂O₃ 0.04%, MgO 0.03%, P₂O₃ 0.007%).

The composition of the invention may contain one or more of the above-described silicarich materials, and, by way of general illustration, it may be said that powders having a composition in the following ranges have been found particularly effective:—

a) 50 to 80% cement and
50 to 20% silica sand;
b) 20 to 55% cement and
45 to 80% shirasu;
c) 20 to 55% cement,
20 to 40% shirasu and
20 to 40% silica sand;
d) 40 to 50% cement,
35 to 47.5% silica sand and
2.5 to 25% zircon flour;
e) 40 to 50% cement,
35 to 48% shirasu and
2.5 to 25% zircon flour; and
f) 40 to 50% cement,
18 to 25% shirasu and
25 to 25% shirasu and

It has also been found that the production of vitreous fibres is in general greatly facilitated by having the silica-rich material of substantially the same particle size as the cement, since this ensures good mixing of the two components when the powder is melted.

2.5 to 25% zircon flour.

The silicon content of the powder used in the process of the invention is preferably

from 45 to 65% (calculated as silica), and most preferably from 50 to 60% (calculated as silica).

The powder is formed into fibres by a spinning operation - that is, it is heated until it is molten, and then passed through orifices or spinnerers to form vitreous fibres. The temperature to which the powder has to be heated so that it melts is, of course, dependent on its composition. It has been found that the spinning operation is most easily performed at a temperature of from 1100 to 1500° C, and thus for convenience the powders should melt well within this range. Powders that melt at temperatures greater than 1500° C give rise to difficulties; they tend to solidify quickly, and this prevents the formation of long fibres. Preferably the powder used in the process of the invention has a melting point of from 1200 to 1300° C.

The process of the invention may be used to prepare either long or short vitreous fibres. By way of illustration only, two spinning apparatuses will now be described with reference to the accompanying drawings, in which:—

Figures 1 and 2 are schematic drawings of apparatuses for producing short and long fibres respectively, which may be employed in the process of the invention.

Figure 1 shows two views of a typical apparatus for producing short fibres; Figure 1A is a front view and Figure 1B is a side view. 95 The powder containing cement and silica-rich material is introduced into the melting furnace (1) where it is heated to a temperature at which it is completely melted. The formed melt flows out through orifices in the bottom 100 of the furnace to form single fibres (2), which are supported by a comb (3), and separated so that none of the fibres cross or become tangled. The fibres are formed into the required thickness by passage between 105 rubber rollers (4), and are then cut to length with a burner (5).

The maximum temperature of the furnace is 1500° C; powders that melt at a temperature higher than 1500° C, such as some powders containing iron, are not used since it is difficult to obtain single fibres. For safety reasons, it is desirable if the furnace is operated at a temperature not higher than 1500° C, and preferably in the range of from 115 1200 to 1300° C.

Figure 2 shows two sides of a typical apparatus for producing long fibres. Figure 2A is a front view and Figure 2B is a side view. The powder containing cement and silica-rich material is introduced into the furnace (1) where it is heated to a temperature at which it is completely melted. The melt is then passed through orifices to form single fibres (2). Several single fibres, to which a binding agent is applied from an applicator (6), are united by a grooved roller (7) to form a strand (8). The formed strand is then wound

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it high speed onto the winding drum (10), the strand being moved across the drum by ineans of a traverse guide (9).

As with the apparatus for producing short fibres, the maximum practical temperature of the furnace in this apparatus is 1500° C.

The following Examples are now given, though only by way of illustration, to show materials, conditions and techniques which 10 may be employed in the process of the invention. In these Examples where mesh sizes are quoted they refer to the JIS standard of mesh sizes.

Example 1.

Production of long vitreous fibres from cement and silica sand.

a) Starting materials

Commercially available portland cement (containing small amounts of gypsum) was formed into a powder with silica sand. Finely powdered (to pass through a 65-mesh sieve) silica-rich material which had been converted into tridymite or cristobalite was employed. Suitable materials are, for example, silica sand from Yuo Island, white clay from Beppu, Japan or commercial silica powder, the chemical compositions of which are shown in the following Table.

Chemical Composition of silica-rich materials

				or stile	a-rich n	aterials			
Silica	SiO,	Al ₂ O,	Fe,O,	TiO,	CaO	MgO	K,0	Na ₂ O	-1
sand from Yuo Island	97.50	0.88	0.014	0.29	0.02	0.10	0.04	0.18	
White Clay			 	 			1	1	1
from Beppu	95.25	1.36	0.12	1.11	0.16				1
Commercial					0.16	0.08	0.02	0.14	
silica powder	98.62	0.51	0.044	0.02	0.07	0.05	0.06	0.03	
Process								0.03	

30 b) Process

The cement/silica powder was first thoroughly mixed in a ball mill and then introduced into a furnace where it was heated to a temperature of 1450° C at which it was 35 completely melted. The formed melt flowed out from nozzles of 4mm in diameter, and the fibres formed were wound onto a drum while being maintained at an appropriate temperature. The fibres obtained were subjected to tests, and the results are sum-

marised in Table 1 below.

When a composition lying outside the range of compositions shown in the Table was used, spinning was impossible at a temperature below 1500°. For example, when the ratio in percentage by weight of the portland cement to the silica powder (selected from the group consisting of white clay from Beppu, Silica sand from Yuo Island and commercial silica powder) was greater than 8/2, the spinning temperature exceeded 1500° C.

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Powder				Cor	Composition					Soing		-	
	SiO	A1,0,	Fe,0,	Ti0,	Ca0	MgO	К,0	Na,0	SO	temperature			Eionga- tion
Ratio of												(kg. mm,)	(2)
cement to													
Silica sand (from Yuo Island)		·											
5:5	29 65	3 0.5	.07								*		
	6:10	2.04	1.60/	0.145	32.61	0.9	0.02	60.0	0.75	1390 - 1440	18.025	698.89	000
6:4	51.96	3.472	1.926	0.116	39.13	1.06	0.016	0.072	06.0	1410	33,000		000
7:3	44 37	3 904	2 244	5000						0441 - 014	23.908	52.278	2.275
			£ 7.7	0.08/	45.646	1.22	0.012	0.054	1.05	1410 - 1440	23.424	59 683	7.5
8:2	36.87	4.336	2.563	0.058	52.164	1.38	0.008	0.036	95			-00.77	55.
Ratio of								0:030	0,7	061 - 0441	20.132	72.400	2.2.25
Portland cement to													-
(from Beppu)								•					
5:5	58.43	1 28	77	0.666									
			7	0.333	32.68	0.89	0.01	0.07	0.75	1390 - 1440	33.533	Car 119	
1 :0	51.06	3.66	1.968	0.444	39.18	1.05	0.008	9500	000	9000		00.203	2.9.20
						7	_	-	⊣	1390 - 1440	29.683	15.460	2.750

VABLE 1 (Continued)

:longa-	
5 - F.	aon I di
r Tensile	strength
Diameter	of fibre
Spinning	temperature
Composition	
Powder	

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Powder			٠.	ပိ	Composition					Spinning	Diameter Tensile of fibre strength	Tensile strength	Elonga- : tron
	SiO2	Al ₁ O,	Fe,0,	Fe,0, Ti0,	Ca0	MgO	MgO K ₂ O Na ₂ O SO,	O, uN	_	(၁၅)	=	(kg/mm,)	(5,)
Ratio of Portland													
cement to Commercial silica powder									,				
\$.5	60.11 2.86	2.86	1.622 0.01	0.01	32.64 0.835 0.03 0.015 0.75	0.835	0.03	0.015	0.75	1440	23.518	43.549	2.500
4:9	52.41 3.32	3.32	1.938		39.128	1.04	0.024 0.012	0.012	06.0	0.90 1420 - 1480	30.310	56.179	2.775
7:3	44.71	3.19	2.253	900.0	2.253 0.006 45.661 1.21	1.21	0.018	0.018 0.009	1.05	1.05 1380 - 1460 19.445	19.445	63.956	1.650
8:2	37.00	37.00 4.26	2.569	0.004	2.569 0.004 52.174 1.37	1.37	0.012	0.006	1.20	0.012 0.006 1.20 1440 - 1450 19.002	19.002	52.728	1.900

Production of long vitreous fibres from cement and shirasu.

a) Starting Material
A powder was prepared by mixing cement and shirasu. The cement used was the same as that used in Example 1. Shirasu is a naturally occurring material, found in large amounts in

Kagoshima Prefecture, Japan, for which, until now, there has been no particularly profitable 10 use, and it is therefore a very inexpensive starting material. The compositions of a number of samples of shirasu are given in Table 2; the shirasu used in this Example had the composition of sample 10, from which 15 any iron was first removed with a magnet.

1446.00
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the state of the s
Ata variatera Brown reviewed matter ad upper part of Ata shirdsau bine time-cho do.06 0.66 15.43 2.82 1.47 0.10 0.87 3.20 1.80 5.8
Att valdeta 17 17 17 17 17 19 19 19
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Polougat low Ata Ata 22 22 22 22 22 22 22 22 22 22 22 22 22
Ata caldera 16 Water-colouned matter at lower shirasu Darjime-cho Vimotsuki-gun 64.67 0.62 15.44 4.72 1.82 0.10 0.98 3.47 3.47 3.47 3.47 3.47 3.47 4.58
Ata cahleta 16 Water-coloune matter at lowe pair of Ata shirasu One jime-cho Kimotsuki-gun 64.67 0.62 15.44 4.72 1.82 0.10 0.98 3.47 3.47 3.47 3.47 0.58
1 1 2 1 2 1 2 1 2 1 2 2 2 2 2 2 2 2 2 2
1 15 15 15 15 15 15 15 15 15 15 15 15 15
Egnchima Higashi Ichiki-cho Hioki-gun Hioki-gun Hioki-gun Hioki-gun Hioki-gun Hioki-gun Hioki-gun Hioki-gun Hioba
Egnehima Higashi Ichiki-cho Hioki-gun Hioki-gun 1.09 1.09 1.41 0.04 0.97 2.90 2.90 83
13 High High High High High High High High
Sakanoue Taniyama city 71.72 0.21 14.14 0.64 1.51 0.05 0.05 0.05 0.48 2.13 3.20 2.13 3.20 2.13
Sakano Taniya City 71.72 0.21 14.14 0.64 0.64 0.05 0.05 0.05 0.05 0.05 0.05 0.05 0.0
11 11 11 11 11 11 11 11 11 11 11 11 11
Aira cho cho
Aira 10 10 Sobegun Sobegun 70.82 0.24 14.02 0.95 1.40 0.07 0.52 1.90 1.30 96 7
Shibushic Soo-gun Soo-gun Soo-gun Soo-gun Soo-gun Soo-gun N0.82 0.24 14.02 0.95 1.40 0.07 0.52 1.90 3.60 0.05
Sanmonji Osaki-cho Soo-gun 71.70 0.20 13.59 0.75 1.47 0.05 0.62 2.07 3.52 2.90 49 100
8 . 86 8 8 1
Aira Shirasu Nakano Kajiki-cho Aira-gun 68.48 0.26 14.74 1.23 1.59 0.37 0.47 0.47 0.47 0.47 0.47 0.47 0.47 0.00 3.53 100.3
S KRAKA KRAK
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b) Process The cement/shirasu powder was introduced into a furnace and heated to a temperature of 1440° C at which it was completely melted. The melt was passed through orifices of 4mm onto a drum while being maintained at an appropriate temperature. The fibres obtained diameter to form fibres, which were wound were tested, and the results are shown in

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ployed having a composition outside the range of composition shown in Table 3, spinning was impossible at a temperature below As in Example 1, if a powder was em-1500° C. Thus, the spinning temperature became higher than 1500° C when the ratio of portland cement to "shirasu" lay outside the range of from 55:45 to 20:80.

TABLE

•				T				_	_	_	_	_			_	_		_	_ 7	Ī	
				\vdash	O'ru	0.5		1.75		1.93		2.10		2.22		2.45		2.74			
				3	2	~ 	:	1.5	;	9.		8.1		<u>6.</u>				~i			
				MeO	2911	16.0		0.85		0.77		89.0		0.60		0.51	;	0.38			1
			c omposition	CaO		37.21		34.10		30.99		27.88		74.77	,	71.00	- 66	76.01		rcon flox	144
		ζ	ا ح	Fe,0,		2.66	- 6	7.60	, , ,	+6.2	-	27:7	-	7+.7	٦٠ ١	7.30	06.6	2		rtures, zi	750. 26
				AI,O,		9.16	0 60	00.	10.01	-	10 48	0.70	10 92		11 36		12.24			above mi	SiO
				SiO,		+4.93	45.30		47.67		50.04	•	52.41	: :	54.78		57.38	1		10 the above mixtures, zircon flour	
		Diameter	of fibre	ĵ.	36031	679.61	15.724		15.904		100.91		14.022		12.588		12.010				
			Elongation	(0)	1.023		1.625		1.598		1.575		1.336		1.075		- CM3		Example 3	Production of long vitreous fibres	
		Tensile	(kg/mm²)		40.167		28.667	763 33	070.66	302 63	27.705	47 533	47.322	41 022	41.000	40 350	000.01		Ero	Production of	from cement siling
		l oad	(8)		17.33		18.05	16 96		15.88		12.01	10:31	7 94	:	8.25			20	,	fro fro
	Admixing ratio	by weight of portland cement	to "shirasu"		55/45	09/03	DC /DC	45/55		10/00		35/65	*	30/70		20/80					

To the above mixtures, zircon flour (weight ratio of SiO₂: ZrO₃ 36:44) which passed through a 300-mesh sieve was added in amounts of 2.5, 7.5, 12.5, 17.5 and 25.0% To the above of the mixture.

Powders were formed containing equal

(i) silica sand from Yuo Island, or amounts (by weight) of cement and (ii) commercial silica powder.

from cement, silica sand and zircon flour.

a) Starting materials

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b) Process
The above powders were thoroughly mixed in a ball mill, and then introduced into a furnace and heated to a temperature of 1490°C at which the mixtures were com-pletely melted. The formed melt was passed

The particle size of each of these powders was such that the powders passed through a

65-mesh sieve.

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through ordices to form fibres, which were wound onto a drum while being maintained at an appropriate temperature.

The fibres obtained were tested, and the results are shown in Table 4.

TABLE 4

such that the maders passed through a 65-mesh sieve. ද

in a ball mill, and the introduced into a temperature of 1490° C at which the mixtures were compictely melied. The formed melt was passed

through orifices to form fibres, which were wound onto a drum while being maintained at an appropriate temperature.

The fibres obtained were tested, and the results are shown in Tuble 4.

		So,	0.74	0.72	0.67	0.74	0.72	0.69
		N.O.	0.10	0.09	0.09 0.08	0.04	6.04	0.04
S.		MgO	0.92	0.83	0.81	0.86	0.84 0.81	0.79
	weight)	CaO	32.13	30.14	27.15	32.15	30.16	29.18
	on (% by	ZrO2	1.00 3.04	5.07	10.14	1.01		7.08
	Composition (% by weight)	Tio ₂	\$1.0 0.14	0.14	6.14	0.01	0.02	0.02
		Fe,0,	1.54	1.49	1.37	1.55	1.50	1.46
TABLE 4		A1,0,	2.90	2.76	2.64	2.74	2.66	
		SiO ₂	58.33	<u> </u>	59.70			
ľ					+-	. 28 	57.19	
-			67.53	71.41	69.42	70.28	82.69	68.17
	Diameter of fibre (μ)	18.43	18.51	19.30	14.14	14.97	15.92	61.71
	Zircon flour	2.5	12.5	17.5 25.0	2.5			0.65
		Cement 50%	Sand (from Yuo Island)		Cement 50% +	Powder 1		

Portland cement containing no gypsum was formed into a powder with from 20 to 40% of shirasu and from 20 to 40% of silica sand. The cement/shirasu/silica powder was introduced into a furnace and heated to a tem-Perature of 1440° C at which it was com-Production of short vitreous fibres from cement, shirasu and silica sand. Example 4. a) Starting material b) Process

25 pletely melted. The formed melt was passed through orifices to form the desired fibres, and these fibres were cut into short lengths c) Results
Physical properties of the short fibres pro-Appearance: White, bulky Diameter: less than 3¹
Length: 5 to 10 mm

0.75

29.67

Content of granular substances: less than

As may be seen from all of the above Examples the process of the present invention makes it possible to form vitreous fibres having excellent characteristics from readily available starting materials.

WHAT WE CLAIM IS:-

1. A process for producing vitreous fibres, in which process a powder comprising from 35 to 75% (calculated as silica) of a silicaceous material, from 10 to 55% (calculated as calcium oxide) of a calciferous material, minor amounts of metal oxides (as hereinbefore defined), and unavoidable impurities, is fused to form a melt which is passed through orifices or spinnerers to form the desired vitreous fibres.

2. A process as claimed in claim 1, in which the silicaceous material is in the form of silicates and/or aluminosilicates.

 3. A process as claimed in claim 1, in which the powder comprises portland cement and a silica-rich material (as hereinbefore defined).

 A process as claimed in claim 3, in which the cement contains substantially no gypsum or other material capable of releasing SO₃.

5. A process as claimed in either of claims 3 and 4, in which the silica-rich material contains greater than 60% (calculated as silica) of silicon.

6. A process as claimed in any of claims 3 to 5, in which the silica-rich material is one or more of silica sand, white clay, shirasu and zirconflour mix.

7. A process as claimed in claim 6, in which the powder contains 50 to 80% cement and 50 to 20% silica sand.

8. A process as claimed in claim 6, in which the powder contains 20 to 55% cement and 45 to 80% shirasu.

9. A process as claimed in claim 6, in

which the powder contains 20 to 55% cement, 20 to 40% shirasu and 20 to 40% silica sand.

10. A process as claimed in claim 6, in which the powder contains 40 to 50% cement, 35 to 47.5% silica sand and 2.5 to 25% zirconflour.

11. A process as claimed in claim 6, in 50 which the powder contains 40 to 50% cement, 35 to 48% shirasu and 2.5 to 25% zirconflour.

12. A process as claimed in claim 6, in which the powder contains 40 to 50% cement, 18 to 25% silica sand, 17 to 25% shirasu and 2.5 to 25% zirconflour.

13. A process as claimed in any of claims 3 to 12, in which the silica-rich material has substantially the same particle size as the cement.

14. A process as claimed in any of the preceding claims, in which the silicon content of the powder is from 45 to 65% (calculated as silica).

15. A process as claimed in claim 14, in which the silicon content is from 50 to 60% (calculated as silica).

16. A process as claimed in any of claims 7 to 12, in which the powder melts in the range of 1100 to 1500° C.

17. A process as claimed in claim 16, in which the powder melts at from 1200 to

18. A process as claimed in claim 1, and substantially as described hereinbefore.

19. Vitreous fibres whenever prepared by a process as claimed in any of the preceding claims.

For the Applicants, SANDERSON & CO., Chartered Patent Agents, 97 High Street, Colchester, Essex.

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20 to 55% 20 to 40% lim claim 6, in 25 to 25% 10 1446910 COMPLETE SPECIFICATION This drawing is a reproduction the Original on a reduced sca. 1 SHEET in claim 6, in 550% cement, 25% zircon-50 claim 6, in 50% coment, chirasu and 55 of claims material has tize as the 60 any of the (calculated 14, in 50 to 60% of claims (8) in the 70 16, in 1200 to I, and 75 red by (B)

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